PATENT SPECIFICATION (11) 1263 221

DRAWINGS ATTACHED

(21) Application No. 9060/70 (22) Filed 25 Feb. 1970
(31) Convention Application No. 15388 (32) Filed 3 March 1969
(31) Convention Application No. 15395 (32) Filed 3 March 1969 in
(33) Japan (JA)
(45) Complete Specification published 9 Feb. 1972
(51) International Classification D 01 f 7/02 7/04
(52) Index at acceptance
B5B 200 350 352 35Y 360 363 365 401 510 720 728 72Y

(54) IMPROVED SYNTHETIC COMPOSITE FILAMENTS

(71) We, TORAY INDUSTRIES, INC., a Company organized and existing under the laws of Japan, of 2, Nihonbashi Muromachi 2-chome, Chuo-ku, Tokyo, Japan, do hereby declare the invention for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

The present invention relates to improved synthetic composite filaments.

Synthetic composite filaments consisting of two or more synthetic polymer materials are well-known, and are for example, as follows:

(1) melt-spun filament which are prepared by melt spinning blend of two or more synthetic polymers, one of which can be dissolved or dispersed in the other,

(2) Conjugate bi-metal-type composite filaments in which two or more component polymers are bonded to each other in a side-by-side relationship along the longitudinal axis of the composite filament,

(3) laminate-type composite filaments in which two or more component polymeric layers are shaped in a tape form or flat form and are mutually laminated,

(4) "core-in-sheath" type composite filaments in which one or more core components are embedded in a sheath component,

(5) modified "core-in-sheath" type composite filaments in which one or more flat core components are embedded in a sheath component and are parallel to each other,

(6) "multi-cores-in-sheath" type composite filaments in which a number of core components are embedded in a sheath component.

The composite filaments of the present invention include the bi-metal type, laminate-type, core-in-sheath type, modified core-in-sheath type and multi-cores-in-a-sheath type composite filaments.

The "multi-cores-in-a-sheath" type composite filaments include "islands-in-a-sea" type composite filaments.

In appearance of a cross-section configuration of the "island-in-a-sea" type composite filament, a number of microfine filamentary core components (island components) are independently embedded in a sheath component (sea component) bonding the core components into a filament body.

Thus, the relation of the core components and the sheath component is illustrated in such a way that the cross-sectional view of the composite filament appears as a number of islands scattered in a sea. In the "islands-in-a-sea" type composite filament, the island components independently extend in the longitudinal direction of the composite filament so as to form a number of micro-fine multi filaments and the sea component fills the spaces between the island components so as to incorporate the micro-fine filaments into a composite filament body and extends in a continuous manner along the longitudinal axis of the composite filament.

In the industrial manufacture of synthetic filaments, the fineness of the filaments which can be obtained is limited because very fine filaments are very difficult to handle without breaking. However, in the case of the composite filaments of the present invention, micro-fine filaments having a very small diameter, which are not obtainable by conventional filament manufacturing processes, can be formed by removing the styrene-copolymeric filamentary component. Particularly, the "islands-in-a-sea" type composite filaments are valuable for manufacturing a bundle of micro-fine filaments.

In practical usage of the "islands-in-a-sea" type composite filament, it is known that the micro-fine filamentary bundle of the island components which is made up from a fibre-forming crystalline polymer can be prepared by removing the sea component of the composite filament with a solvent suitable for dissolving the sea component. The bundle of the micro-fine filamentary island

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components is useful for producing valuable sheet material. It is also known that the removing operation may be carried out after forming the composite filaments into a desired form such as yarn, knitted fabric, woven cloth, sheet, mat, non-woven material, and other textile materials.

When the composite filaments or these products are subjected to the removing operation for producing the micro-fine filamentary bundle stated above, it is desirable that the component to be removed, such as the sea component, satisfies the following requirements;

(a) low cost,
(b) easy dissolution,
(c) high productivity of the composite filament, and
(d) easy processing, such as spinning into yarn, knitting, weaving and non-woven material forming, of the resultant composite filaments.

Styrene polymers can be used for obtaining the component to be removed satisfying the above-described requirements, because the styrene polymers may be easily dissolved with relatively low cost solvents such as tri-chloroethylene, tetrachloroethylene, aromatic hydrocarbons such as toluene and xylene, chlorinated carbons such as carbon tetrachloride, dimethyl acetamide, dimethyl sulfoxide, and dimethyl formamide. Needless to say, in the dissolving of the component to be removed by a solvent, it is required that the components of the composite filament remaining are never dissolved by the solvent.

Fiber forming polyamides such as nylon 6, nylon 66, nylon 12, nylon 610, nylon 11, block polyetheramides, copolymers and mixtures of these polyamides; fiber forming linear polyesters such as polyethylene terephthalate; polypropylene terephthalate, polybutylene terephthalate and polyoxyethylene benzoate are not dissolved in the above-mentioned solvents for styrene polymer, in general.

Also, crystalline polyolefins such as polyethylene, polypropylene, these copolymers and these mixtures are not dissolved in the above-mentioned solvent at room temperature, in general. Therefore, the styrene polymers and the above-mentioned fiber formable crystalline polymers are suitable for production of the composite filament for obtaining micro-fine filaments in which the styrene polymers form a component to be removed by a solvent and the fiber formable crystalline polymers form components to be formed into micro-fine filaments.

For example, in the case of the "islands-in-a-sea" type composite filaments, the styrene polymers form a sea component and the crystalline polymers form a plurality of island components. However, such composite filaments containing the styrene polymer component have the following disadvantages:

1. On the resultant composite filament,
   (a) very brittle, thus, easily breakable,
   (b) high frictional resistance,
   (c) low bending strength,
2. Upon processability of the resultant composite filament,
   (a) difficult opening of fibers,
   (b) easily forming of undesirable pills in the carding step,
   (c) undesirable over-bulkiness of resultant web,
   (d) non-uniform density distribution of resultant web,
   (e) frequently producing of needle breakage in needle-punching operation,
   (f) low efficiency of needle-punching,
   (g) difficult forming of high density sheet,
   (h) easy marking of undesirable wrinkles which is the same as wrinkles created on paper by bending action,
   (i) Easy creation of a number of unfavourable crackings on the resultant filament surface in drawing and/or crimping process, the crackings cause increase of undesirable unevenness and undesirable whiteness of the surface, and;
3. On the resultant products,
   (a) easily creating undesired bending or folding marks,
   (b) easily creating undesired wrinkles,
   (c) low density, and
   (d) undesirable creaky hand feeling.

Owing to the disadvantages stated above, when the composite filament consisting of styrene polymer component and fiber formable crystalline polymers component are subjected to carding or needle punching process in order to produce webs or non-woven fabrics, the styrene polymer-component tends to be frequently broken and causes separation of the crystalline polymer component from the styrene polymer component by mechanical action of the processings.

In the case of the "islands-in-a-sea" type composite filament, the island components separated thus, create numerous pills or neps in the web or non-woven fabric. Particularly, in the case where a non-woven fabric is manufactured from a web of the composite filaments through a needle punching process, the outer portions of the island components, which are exposed outside the composite filament and fibrilized, create a number of neps which cause the insufficient uniformity of the resultant web and breaking of the needle by catching the neps by the barb of the needle, and movement of the individual composite filament in the web accompanied by the needle punching is greatly suppressed and followed by breaking of the composite filament owing to the high frictional resistance and brittleness thereof.

Therefore, products from the conventional composite filaments, particularly the conventional "islands-in-a-sea" type composite filaments consisting of the crystalline polymer island components and a styrene polymer sea component have undesirable creaky hand feeling, insufficient recoverability from dimensional deformation and insufficient uniformity.

In order to overcome the above-stated disadvantages of the conventional composite filament containing the styrene polymer component, many improvements were provided. That is, in order to reduce the brittleness and the frictional resistance of the polystyrene sea component, polyethylene glycol was blended with the polystyrene sea component, a large amount of oiling agent was applied to the composite filament surface, or in order to prevent the polystyrene sea component from breaking, nylon polymer was blended with the polystyrene sea component. However, the improvements stated above were unsatisfactory for overcoming the disadvantages.

According to the present invention, a synthetic composite filament comprises a first filamentary component consisting of a copolymer of styrene with acrylonitrile and/or methyl methacrylate or a mixture of such copolymers, and a second filamentary component consisting of a synthetic crystalline fibre-forming polymer, the first and second components extending along the length of the composite filament, respectively and being bonded together continuously along the length of the composite filament.

An additional copolymerization unit effective for increasing the solubility of the styrene-copolymeric component in the organic solvents as indicated hereinafter may be selected from the vinyl monomers such as methyl styrene, α-methyl styrene, ethyl styrene, dimethyl styrene, chlorostyrene and p-dimethyl aminostyrene, p-methoxy styrene, vinyl pyridine, chloro-α-methylstyrene, N,N-diphenyl acrylamide, p-nitro-styrene and α-phenyl-vinylacetate, fumaronitrile, maleinitrile, 1,2-dicyano-1-methylethylene, 1,2-dicyano - 1 - chloroethylene, -1,2 - dicyano-1 - phenylethylene, β-cyano acrylic esters, α - phenyl - vinyl acetate, N,N - diphenyl acrylamide, vinyl carbazole, butadiene, maleic acid anhydride, vinyl acetate, vinyl alcohol, vinyl chloride, vinylidene chloride, methyl acrylate, ethyl acrylate, butyl acrylate, vinyl pyrolidone, diethyl maleate, diethyl fumalate, butyl methacrylate and methacrylamide. These additional copolymerization units are included with a relative smaller content in the styrene-copolymer so as to never obstruct the purpose of the present invention. If the styrene-copolymer in the first component consists of two components namely acrylonitrile and styrene, the content of acrylonitrile in the copolymer is 10 to 40% by weight, further preferably 10 to 24%.

It is well-known that a mixture of 24% by weight of acrylonitrile and 76% by weight of styrene is copolymerized to an azeotropic copolymer which has a composition the same as that of the mixture.

Therefore, it is preferable to produce a copolymer having a constant composition where the content of acrylonitrile in the acrylonitrile-styrene copolymerization system is 24% by weight. The acrylonitrile unit in the acrylonitrile-styrene copolymer is effective for increasing the resistance for thermal decomposition at a high temperature, and thus, the copolymer is effective for easily processing melt-spinning of the composite filament at a high temperature and for obtaining a high tenacity composite filament. If the content of acrylonitrile in the copolymer is lower than 10% by weight, the resultant composite filament has a tendency to create many pills during carding and breaking of many needles during needle punching. On the other hand, if the content of acrylonitrile in the copolymer is higher than 40%, the solubility of the resultant sea component in organic solvent, as indicated hereinbefore, is unsatisfactory, and processing of spinning for fiber forming becomes difficult.

Where the copolymeric component is a copolymer of styrene and methyl methacrylate, the content of methyl methacrylate is within the range of 10% to 90% by weight; more preferably, the content of methyl methacrylate is 50% to 90% by weight and the content of styrene is 50% to 10% by weight.

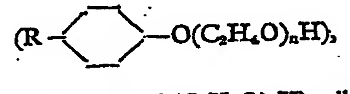
The composite filament comprising the styrene-methyl methacrylate copolymer as the first filamentary component has an excellent transparency, high bright whiteness and high sheen.

If the content of methyl methacrylate in the copolymer is lower than 10% by weight, the purpose of the present invention is insufficiently achieved. If the content of the methyl methacrylate is higher than 90% by weight, the resultant first filamentary component in the composite filament has an insufficient spinnability and unsatisfactory solubility in the solvents.

Where the first filamentary component is prepared from the copolymer consisting of styrene, acrylonitrile and methyl methacrylate, the content of styrene is at least 10% by weight, the content of acrylonitrile is at most 10% by weight and the content of methyl methacrylate is greater than 10% and smaller than 90%. The acrylonitrile unit for copolymerization is effective for increasing the resistance for thermal decomposition at a high temperature of the resultant copolymer, and for obtaining an easy spinnable and drawable composite filament.

If the composition of the styrene-acrylonitrile-methyl methacrylate copolymer does not satisfy the above-mentioned definition, the following disadvantages are encountered, namely in the resultant copolymer, a low resistance for thermal decomposition at a high temperature and a low solubility in the solvents; in the resultant composite filament, a low transparency, an undesirable yellowing and a low sheen; and in the processing of fiber forming, a low spinning ability and a low drawing ability.

The styrene-copolymeric component of the composite filament according to the present invention may contain polyethylene glycol, polypropylene glycol, ethylene glycol-propylene glycol copolymer, or their derivatives, such as alkyl-phenol ethers



alkyl ethers ($R-O(C_2H_4O)_nH$) alkyl amines, alkyl amides, alkylthioethers, phosphoric esters and fatty acid amide and the like in order to improve the frictional property of the composite filament.

The crystalline polymeric component of the composite filament according to the present invention may be made up from a fiber-forming crystalline polyamide such as nylon 6, nylon 66, nylon 610, nylon 12, nylon 11, nylon 9, block polyetheramide, other fiber-forming linear polyamides such as polyamides including the cyclohexane group and polyamides including isophthalic acid, phthalic acid or terephthalic acid, and these copolymers and blends.

Also, the second polymeric component of the composite filament according to the present invention may be a linear polyester which consist of at least one acid polycondensation unit selected from the group consisting of terephthalic acid, isophthalic acid, phthalic acid, p-hydroxy benzoic acid, adipic acid, sebacic acid, trimellitic acid, pyromellitic acid, and sodium sulfoisophthalic acid and at least one glycol polycondensation unit selected from the group consisting of ethylene glycol, propylene glycol, butylene glycol, cyclohexane dimethanol and diethylene glycol, and mixtures of two or more of these units.

Furthermore, the second polymeric component of the composite filament according to the present invention may be formed from a fiber-forming crystalline polyolefin such as polyethylene, polypropylene, copolymers of ethylene and propylene and mixtures of these polymers.

The present invention will be further described with reference to the accompanying drawings, in which;

Figs. 1A to 1F are cross-sectional model views of several embodiments of the composite filaments of the present invention,

Fig. 2A is a partially sectional model view of an embodiment of the composite filament of the present invention,

Fig. 2B is a side model view of another embodiment of the composite filament of the present invention,

Fig. 3 is a cross-sectional view of an embodiment of a spinneret for spinning the composite filament of the present invention,

Fig. 4 shows a range of draw ratios suitable for drawing undrawn composite filaments to form composite drawn filament of the present invention.

In Figs. 1A to 1G, a plurality of island components 1 are embedded within a sea component 2. Fig. 1A shows a typical "islands-in-a-sea" type composite filament in which the island components 1 are arranged within the sea component 2 in a predetermined order. Fig. 1B shows a modified composite filament in which, some of the island components 1 are exhibited on the outside surface of the composite filament. Fig. 1C shows an embodiment of the composite filament in which a number of very fine island components 1 are dispersed in the sea component 2.

In the composite filament shown in Fig. 1D, two or more island components are closely incorporated into a filamentary body along the longitudinal axis of the composite filament. Fig. 1E shows a composite filament in which the island components are arranged in a flowery order. Fig. 1F shows a modified composite filament having a hollow cross-sectional profile. Fig. 1G shows an irregular composite filament having a triangular cross-sectional profile. The composite filament of the present invention may be any irregular filament such as a filament having a T-shaped or Y-shaped cross-sectional profile.

Fig. 1H shows a specific composite filament which is an assembly of a plurality of bi-metal type filamentary elements in which a crystalline polymeric filamentary component 1 and a styrene-copolymeric filamentary component 2 are incorporated side-by-side into a filament body.

The distribution of the island components in the "islands-in-a-sea" type composite filament in the cross-sectional view may be uniform or random but a uniform distribution is desirable in order to produce a microfine filamentary bundle with high efficiency. The arrangement of the island components in the cross-sectional view may be a β-arrangement in which the island components are arranged in a single circular order along the peripheral surface of the composite filament, or an α-arrangement in which the island components are arranged so that one or more island components are surrounded with the island components arranged in accordance

with the β-arrangement. However, the α-arrangement is desirable for the composite filament of the present invention in order to obtain softer sheet products.

Further, in order to obtain "island-in-a-sea" type composite filaments useful for processing through the carding and/or needle-punching process with high efficiency, it is preferable that the island components are completely surrounded by the sea component so that the island components are not exposed on the peripheral surface of the filament. Therefore, the composite filaments having the cross-sectional profile as indicated in Figs. 1A, 1C, 1E and 1G are suitable for this purpose.

In Fig. 2A, the micro-fine filamentary island components 1 are independently embedded within the sea component 2 in parallel along the longitudinal axis of the composite filament. Fig. 2B shows a longitudinal cross section of the composite filament of Fig. 1D, where the island components 1 and 1a, positioned at the outside portion of the composite filament, are exposed on the peripheral surface of the composite filament and independently extended in parallel along the longitudinal axis of the composite filament.

If a fiber-forming polymer formed in pellet-form is used for melt-spinning at a high temperature, it is desirable to dry the polymer pellet in a vacuum condition at a temperature lower than the plasticizing point of the polymer. In such drying, it is required that the polymer pellet is protected from contact with water, oxygen and air in order to protect the polymer from decomposition. But, in case the styrene-copolymer is used for the melt-spinning in pellet form in order to form the filament, the above-stated protections for the styrene-copolymer are not necessarily required. If necessary, addition of a small amount of stabilizer, such as anti-oxidant, is very effective as the preventative. In melt-spinning for the composite filament of the present invention, a spinning chimney for maintaining the filaments spun through a spinneret at a desired constant temperature is effective for improving of extensibility of undrawn filaments, and tenacity and elongation of the resultant composite filament.

Fineness and length of the composite filament of the present invention may be adjusted at will according to the intended use of the composite filament. Generally, the fineness is adjusted in a range from 1.5 to 20 denier, preferably from 3 to 10 denier. Fineness of the micro-fine filament, which is obtained from the island components of the composite filament, is from 0.005 to 1.0 denier, preferably, from 0.01 to 0.5 denier. The number of the micro-fine filaments to be obtained from the composite filament may be adjusted at will, preferably, the number being 10 or more.

In the case where the sea component is removed from the composite filament by a solvent treatment, it is desirable that content of the crystalline polymeric components such as island components in the "islands-in-a-sea" type composite filament be as high as possible, practically, from 40 to 75% based on the weight of the composite filament, and fineness of the micro-filamentary crystalline polymeric component is from 0.01 to 0.5 denier. If the fineness and content of the filamentary crystalline polymeric component is lower than 0.01 denier and higher than 75% respectively, the crystalline polymeric components tend to create an undesirable adhering of the crystalline polymeric components with an adjacent one.

The "islands-in-a-sea" type composite filament of the present invention may be manufactured by means of a specified spinning apparatus, for example, as indicated in Fig. 3. In Fig. 3, three kinds of spinnerets 4, 5 and 6 are contained within a spinning device 3 in combination. A partition 7 is usable for independently feeding a sea component polymer B and an island component polymer A into the spinnerets 5 and 6, respectively.

The spinnerets 4 and 5 are provided with a plurality of orifices, respectively. The lower ends of the orifices 11 of the spinneret 4 are inserted into the upper ends of the orifices 12 of the spinneret 5. The molten sea component polymer B is fed into the orifices 12 of the spinneret 5 through a passage 9 and then fed into spaces between the lower ends of the orifices 11 of the spinneret 4 and the upper ends of the orifices 12 of the spinneret 5. The molten island component polymer A is fed into the orifices 11 of the spinneret 4 through passages 8 connected with the orifices 11 of the spinneret 4, and further fed into the orifices 12 of the spinneret 5. Through contacting of the molten polymers A and B in the spinneret 5, both polymers A and B form a composite stream in which the polymer B surrounds the polymer A having an approximate circular cross-sectional profile.

The spinneret 6 is provided with a plurality of orifices 13 and funnel-shaped chambers 10, an upper end of which connects with lower ends of the orifices 12 of the spinneret 5 and a lower end of which connects with the orifices 13 of the spinneret 6.

A plurality of the composite streams of the molten polymers A and B are fed into the funnel-shaped chambers 10 through the orifices 12 of the spinneret 5, and incorporated into an "island-in-a-sea" type composite stream.

An "islands-in-a-sea" type undrawn composite filament is obtained from the "island-in-a-sea" type composite stream of the molten polymers A and B.

In order to prepare the above-mentioned

"assembled-bi-metal" type composite filament as indicated in Fig. 1D or 1H, a plurality of the composite streams in which a molten polymer stream A and a molten polymer stream B are combined in a bi-metal form, are fed into the funnel-shaped chamber 10 through a conventional orifice for spinning the bi-metal type composite filament, for example, the orifice indicated in British Patent No. 1,171,843, and incorporated into an "assembled-bi-metal" type composite stream.

The undrawn composite filament is preferably drawn at a relatively low draw ratio. For example, for an "islands-in-a-sea" type composite filament composed of a sea component of acrylonitrile-styrene (24:76) copolymer and 16 island components of nylon 6, an undrawn composite filament drafted at a take-up velocity of 1500 m/min is preferably drawn in a draw ratio of 1.5 to 3.0, and an undrawn filament taken up in a velocity of 1000 m/min is preferably drawn at a draw ratio of 2.0 to 3.4. A range of drawing ratios suitable for drawing the undrawn composite filament of the present invention is illustrated in Fig. 4 in a relation with respect to the taking up velocity for preparing the undrawn composite filament. In Fig. 4, the proper draw ratio for preparing the composite filament according to the present invention may be selected from the shaded portion of this graph based upon the take-up velocity of the composite filament. If the draw ratio is higher than the upper limit of the shaded range, the separation tendency of the island components from the sea component is undesirably increased.

And, if the draw ratio is lower than the lower limit of the shaded range, the tenacity of the resultant composite filament is insufficient.

The staple fibers prepared from the composite filaments of the present invention are useful for forming a web by the conventional machine such as card, cross-lapper, random-webber for the staple fibers and a sheet forming machine for endless filaments. The composite staple fibers of the present invention may be mixed with another kind of fiber for forming the web. The web thus obtained may be subjected to a needle-punching process for producing sheet materials with a felting needle selected from the needles having a wider range of thickness and length than the felting needle used for the conventional web. The web may be needle-punched under a condition in which the web is laminated with the woven or knitted fabric or non-woven cloth.

The styrene-copolymeric component in the composite filament may be removed by dissolving off with a solvent such as trichloroethylene, tetrachloroethylene, toluene, xylene, benzene, acetone, methylethyl ketone, tetrachloromethane, dimethyl acetamide, dimethyl sulfoxide, and dimethyl formamide, in a film-form, web-form or fabric-form.

The composite filament of the present invention is valuable for manufacturing the various products such as artificial leather, carpet, filter and mopping cloth by way of the following conventional processes:

(a) web forming — sheet material preparing — dissolving of sea component — product

(b) web forming — sheet material preparing — sizing with a water soluble polymer — impregnation of an elastic polymer solution — coagulation of the elastic polymer desizing and dissolving of sea component — product

(c) web forming — sheet material preparing — sizing with a water soluble polymer — dissolving. of sea component — impregnation of an elastic polymer solution — coagulation of the elastic polymer — desizing — product

(d) web forming — sheet material preparing — dissolving of sea component — impregnation of an elastic polymer solution — coagulation of the elastic polymer — product

(e) web forming — sheet material preparing — dissolving of sea component — sizing — impregnation of an elastic polymer solution — coagulation of the elastic polymer — desizing — product.

The composite filament of the present invention, which is characterized by comprising a styrene-copolymeric filamentary component which is easily soluble into the low cost solvent such as trichloroethylene and tetrachloroethylene, and a fiber formable crystalline polymeric filamentary component, has the following advantages;

(1) easily web-forming and needle-punching for forming a web owing to high resistance of the styrene-copolymeric filamentary component against its breaking,

(2) uniform intertwining of the filaments,

(3) not creating the so-called "corrugated board structure" in the resultant web and sheet material,

(4) high density of the resultant web and sheet material,

(5) high resistance against peeling of the web,

(6) high resistance aaginst the creating of bending marks and wrinkle marks, and

(7) easily dissolving of the sea component with a solvent.

The present invention is further illustrated by the following example 8 in which the proportions of the ingredients are expressed as parts by weight and which are given for illustrative purposes only.

Example 1

This is relative to the preparation of an "islands-in-a-sea" type of composite filament.

Sea component polymer pellets were prepared from a copolymer of 24% acrylonitrile and 76% styrene by means of a melt-

extruder with a cutter. Island component polymer pellets were prepared from nylon 6 polymer in the same manner. Both polymer pellets were dried and fed to a melt extruder in equal parts by weight and the polymer melts were extruded at 285°C by means of the melt-spinning shown in Fig. 3 to form an "islands-in-a-sea" type undrawn composite filament having the cross-sectional profile shown in Fig. 1A, and taken up at a speed of 1500 m/min through a quench chamber and spinning chimney and oiling bath. The spinning process was carried out under fovourable condition. The thus obtained undrawn filaments were drawn using a heating pin maintained at 85°C and a heating plate maintained at 125°C at a ratio of 2.25. The drawn filament had a fineness of 5.1 deniers, therefore, each of 16 island component filaments in the composite filament had a fineness of 0.16 deniers. The drawn filaments were doubled and crimped to 12 crimps/inch by a crimper, and cut to a length 51 mm by means of a cutter to form staple composite fibres.

The resultant "islands-in-a-sea" type composite staple fibres were formed into a uniform cloth lap without difficulty by passing them through a carding machine and a lap machine.

The cloth lap was smoothly subjected to a needle punching process at a needling condition of 500 needles/cm² without any needle breaking. Through the needle punching, the cloth lap was favourably converted to a felt sheet having a weight of 300 g/cm².

The felt sheet was impregnated with a solution of 10% of polyvinyl alcohol in water and then dried. The dried felt sheet was treated by spraying trichloroethylene on to the felt sheet in order to dissolve off the sea component consisting of the styrene-acrylonitrile copolymer, squeezed by means of a mangle and then dried. A sheet consisting of bundles of the island component filaments was obtained. The sheet was impregnated with a solution of polyurethane in N,N-dimethyl formamide. After the impregnation step the polyurethane was coagulated by passing the sheet through an aqueous coagulating bath, the sheet was then treated with hot water at a temperature from 95 to 98°C in order to dissolve the polyvinyl alcohol out of the sheet, and then dried. A soft sheet was obtained. The sheet was buffed, sheared and then softened with a softening machine. A very soft leather-like sheet was obtained. The sheet had a high bending strength (over a million times of bending at a temperature of −5°C), a high moisture permeability and an appearance and touch similar to those of a velour-like natural leather.

Example 2

"Islands-in-a-sea" type composite filaments having a cross-section profile as shown in Fig. 1A, were prepared in the manner described in Example 1, except that the island component was nylon 66 polymer, the sea component was a copolymer of 18% acrylonitrile and 82% styrene, and the number of islands components was 24.

The resultant undrawn filaments and drawn filaments were subjected to the following testing.

(1) Test of drawing ability of undrawn filament.

An undrawn composite filament spun and taken up at a velocity of 1500 m/minute was drawn at a draw ratio of 3.2 by using a heating pin at a temperature of 100°C and a heating plate at a temperature of 125°C. The resultant drawn filament was estimated on degrees of sheen, evenness of the surface and whiteness thereof.

(2) Test for carding property of drawn filament.

The drawn and cut composite filaments were subjected 50 times to a hand carding test or once to practical carding, and formed into a web. Processability of the carding was estimated.

(3) Test for needle punching property of web.

A web prepared from the cut composite filament was subjected to needle punching with 500 needles at a needling condition of 100 to 1,000 needles/cm².

Estimation for the test was carried out by consideration of number of needles broken during the punching process and of density of the resultant felt sheet.

(4) Pilling Test

The resultant felt sheet of the needle punching test was estimated by consideration of the pills formed thereon.

(5) Dissolving test for sea component

The drawn composite filaments comprising 3 g of sea component were immersed into 300 ml of trichloroethylene at room temperature. The time for complete dissolving off of the sea component was determined for estimating the dissolving velocity of the sea component.

The resultant undrawn composite filaments of the present Example had a superior drawing ability.

The resultant drawn composite filaments had excellent carding and needle punching properties. The resultant felt sheet had no pills thereon. Further, the sea component of the resultant composite filament was dissolved off at a dissolving velocity similar to that of polystyrene.

For comparison, a composite filament con-

sisting of nylon 6 island components and a polystyrene (commercial grade) sea component was prepared in the same manner as indicated above. The comparison resulted in a drawn composite filament having an uneven and rough filament surface and an undesirable whited appearance, a drawn composite filament having poor carding and needle punching properties, and a felt sheet having a number of pills thereon.

EXAMPLE 3

The procedure of Example 1 was repeated using a nylon 12 island component and a sea component consisting of a copolymer of 22% acrylonitrile and 78% styrene.

The resultant undrawn composite filaments had a smooth surface, and drawn filaments had excellent carding and needling properties under the same conditions as described in Example 2. Also, the felt sheet prepared from the filaments had no pills thereon, and the sea component was easily dissolved off by trichloroethylene at a similar velocity to that of polystyrene.

Comparison composite filaments prepared from nylon 12 island component and polystyrene (high fluidity grade) sea component had poor carding and needling properties, and the felt sheet from the comparison filaments had a number of pills thereon.

EXAMPLE 4

The procedure of Example 1 was repeated using a nylon 610 island component and a sea component consisting of a copolymer of 20% acrylonitrile and 80% styrene.

When tested as described in Example 2, the resultant undrawn composite filaments had smooth surface, and drawn filaments had excellent carding and needling properties. Also, the felt sheet prepared from the filaments had no pills thereon, and the sea component was easily dissolved off by trichloroethylene at a similar velocity to that of polystyrene.

Comparison composite filament prepared from nylon 610 island component and polystyrene sea component had poor carding and needling properties, and the felt sheet from the comparison filaments had a number of pills thereon.

EXAMPLE 5

The procedure of Example 1 was repeated using a nylon 6 island component and a sea component consisting of a copolymer of 15% acrylonitrile and 85% styrene.

When tested as described in Example 2, the resultant undrawn composite filaments had a smooth surface, and drawn filaments had excellent carding and needling properties. Also, the felt sheet prepared from the filaments had not any pills thereon, and the sea component was easily dissolved off by trichloroethylene at a similar velocity to that of polystyrene.

Comparison composite filament prepared from nylon 6 island component and polystyrene (high fluidity grade) sea component had poor carding and needling properties, and the felt sheet from the comparison filaments had a number of pills thereon.

EXAMPLE 6

The procedure of Example 1 was repeated using a nylon 66 island component and a sea component consisting of a copolymer of 12% acrylonitrile and 88% styrene.

When tested as described in Example 2, the resultant undrawn composite filaments had a smooth surface, and drawn filaments had excellent carding and needling properties. Also, the felt sheet prepared from the filaments had no pills thereon, and the sea component was easily dissolved off by trichloroethylene at a similar velocity to that of polystyrene.

A nylon 66 island component and a polystyrene sea component having a high polymerization degree failed to prepare comparison composite filaments owing to poor spinnability thereof.

EXAMPLE 7

The procedure of Example 1 was repeated using a nylon 6 island component and a sea component consisting of a copolymer of 10% acrylonitrile and 90% styrene.

The resultant undrawn composite filaments had a smooth surface, and drawn filaments had excellent carding and needling properties at the same conditions as indicated in Example 2. Also, the felt sheet prepared from the filaments had no pills thereon, and the sea component was easily dissolved off by trichloroethylene at a similar velocity to that of polystyrene.

EXAMPLES 8 TO 11

The procedure of Example 1 was repeated four times using 60 parts island component polymer and 40 parts sea component styrene-copolymer, respectively, instead of the island and sea components of Examples 1 to 4.

When tested as described in Example 2, each of the resultant undrawn composite filaments had a smooth surface, and drawn filaments had excellent carding and needling properties. Also, the felt sheet prepared from each of the filaments had no pills thereon, and each of the sea components was easily dissolved off by trichloroethylene at a similar velocity to that of polystyrene.

EXAMPLES 12 AND 13

The procedure of Example 1 was repeated twice using 30 parts island component nylon pellet and 70 parts sea component styrene copolymer pellet, respectively, instead of the

island and sea components of Examples 1 and 2 in order to prepare composite filaments as indicated in Fig. 1C.

When tested as described in Example 2, each of the resultant undrawn composite filaments had a smooth surface, and drawn filaments had excellent carding and needling properties. Also, the felt sheet prepared from each of the filaments had no pills thereon, and each of the sea components was easily dissolved off by trichloroethylene at a similar velocity to that of polystyrene.

EXAMPLE 14

The procedure of Example 1 was repeated using island components consisting of a copolymer of 85% nylon 6 and 15% nylon 66 instead of nylon 6 used in Example 1.

The resultant undrawn composite filaments had a smooth surface, and drawn filaments had excellent carding and needling properties at the same conditions as indicated in Example 2. Also, the felt sheet prepared from the filaments had no pills thereon, and the sea component was easily dissolved off by trichloroethylene at a similar velocity to that of polystyrene.

EXAMPLE 15

The procedure of Example 1 was repeated, except that the island component was polyethylene terephthalate, and the drawing of the undrawn filament was carried out at a temperature of 165°C in a superheated steam atmosphere. The cross-sectional profile of the resultant composite filament was as shown in Fig. 1G.

The resultant undrawn composite filaments had a smooth surface, and drawn filaments had excellent carding and needling properties under the conditions described in Example 2. Also, the felt sheet prepared from the filaments had no pills thereon, and the sea component was easily dissolved off by trichloroethylene at a similar velocity as that of polystyrene.

Comparison composite filaments prepared from polyethylene terephthalate island component and polystyrene (high fluidity grade) sea component had poor carding and needling properties, and the felt sheet from the comparison filaments had a number of pills thereon.

EXAMPLE 16

The procedure of Example 15 was repeated, except that the island component was polypropylene terephthalate.

The resultant undrawn composite filaments had a smooth surface, and drawn filaments had excellent carding and needling properties under the conditions described in Example 2. Also, the web prepared from the filaments had no pills thereon, and the sea component was easily dissolved off by trichloroethylene at a similar velocity to that of polystyrene.

EXAMPLE 17

The procedure of Example 1 was repeated, except that the island component was polypropylene, 40 parts sea component styrene-copolymer and 60 part island component polymer were applied to spinning, and the resultant composite filament had the cross-sectional profile of Fig. 1E.

The resultant undrawn composite filaments had a smooth surface, and drawn filaments had excellent carding and needling properties under the conditions described in Example 2. Also, the felt sheet prepared from the filaments had no pills thereon, and the sea component was easily dissolved off by trichloroethylene at a similar velocity to that of polystyrene.

EXAMPLE 18

The procedure of Example 17 was repeated, except that the island component was high density polyethylene.

The resultant undrawn composite filaments had a smooth surface, and drawn filaments had excellent carding and needling properties under the conditions described in Example 2. Also, the web prepared from the filaments had no pill thereon, and the sea component was easily dissolved off by trichloroethylene at a similar velocity to that of polystyrene.

EXAMPLE 19

The procedure of Example 1 was repeated, except that the sea component was a copolymer of 45% methyl methacrylate and 55% styrene, and the cross-sectional profile of the resultant composite filament was that of Fig. 1F.

The resultant undrawn composite filaments had a smooth surface, and drawn filaments had a high transparency, a bright sheen, and excellent carding and needling properties under the conditions described in Example 2. Also, the felt sheet prepared from the filaments had no pills thereon, and the sea component was easily dissolved off by trichloroethylene at a similar velocity to that of polystyrene.

The felt sheet from the composite filament of the present Example was valuable for manufacturing synthetic leather having an appearance and touch similar to those of natural leather by the processes of Example 1.

EXAMPLE 20

The procedure of Example 1 was repeated, except that the island component was polyethylene terephthalate and the sea component was a copolymer of 70% of methyl methacrylate and 30% of styrene.

The resultant undrawn composite filaments had smooth surface, and drawn filaments had a high transparency, a bright sheen and excellent carding and needling properties under

the conditions described in Example 2. Also, the prepared from the filaments had not any pills thereon, and the sea component was easily dissolved off by trichloroethylene at a similar velocity to that of polystyrene.

The preparation of comparison composite filament from polyethylene terephthalate island component and polyethyl methacrylate sea component was failed owing to poor spinnability thereof.

EXAMPLE 21

The procedure of Example 1 was repeated, except that the sea component was a copolymer of 72% of methyl methacrylate, 24% of styrene and 4% of acrylonitrile, the spinning was carried out at a temperature of 280°C, the solidification of the spun filaments was carried out by cooling with ambient air without blowing of cooling air, the drawing was carried out at a draw ratio of 2.5, the undrawn filament fed from a feed roller contacting the peripheral surface of a titanium pin which was disposed above a fish tail spray device and heated by steam at a temperature of 100°C supplied from the spray device, being wound on the pin, and then being passed through a steam box to which superheated steam at a temperature of 130°C was supplied. The spinning and drawing processes were carried out without difficulty. The resultant filament had a smooth surface, a high transparency and a fineness of 5.6 deniers, so that each of the 16 island component filaments had a fineness of 0.175 deniers.

The resultant drawn filaments had excellent carding and needling properties under the conditions described in Example 2. Also, the felt sheet prepared from the filaments had no pills thereon, and the sea component was easily dissolved off by trichloroethylene at a similar velocity to that of polystyrene.

EXAMPLE 22

The procedure of Example 21 was repeated, except that the island component was nylon 66, the sea component was a copolymer of 70% of methyl methacrylate, 26% of styrene and 4% of acrylonitrile.

The resultant undrawn composite filaments had a smooth surface, and drawn filaments had excellent carding and needling properties under the conditions described in Example 2. Also, the felt sheet prepared from the filaments had no pills thereon, and the sea component was easily dissolved off by trichloroethylene at a similar velocity to that of polystyrene.

However, the preparation of comparison composite filaments from island component nylon 66 polymer and sea component polymethyl methacrylate failed owing to poor spinnability thereof.

EXAMPLE 23

The procedure of Example 21 was repeated, except that the island component was nylon 610, the sea component was a copolymer of 72% of methyl methacrylate, 22% of styrene and 6% of acrylonitrile, and 40 parts sea component styrene-copolymer and 60 parts island component nylon 610 polymer were applied to spinning.

The resultant undrawn composite filaments had smooth surface, and drawn filaments had excellent carding and needling properties under the conditions described in Example 2. Also, the felt sheet prepared from the filaments had no pills thereon, and the sea component was easily dissolved off by trichloroethylene at a similar velocity to that of polystyrene.

EXAMPLE 24

The procedure of Example 21 was repeated, except that the island component was a mixture of 80 parts nylon 6 and 20 parts nylon 66, and the sea component was a copolymer of 60% of methyl methacrylate, 35% of styrene and 5% of acrylonitrile.

The resultant undrawn composite filaments had smooth surface, and drawn filaments had excellent carding and needling properties under the conditions described in Example 2. Also, the belt sheet prepared from the filaments had no pills thereon, and the sea component was easily dissolved off by trichloroethylene at a similar velocity to that of polystyrene.

EXAMPLE 25

The procedure of Example 21 was repeated, except that the island component was nylon 12, the sea component was a copolymer of 50% of methyl methacrylate, 44% of styrene and 6% of acrylonitrile, and 30 parts of sea component styrene-copolymer and 70 parts of island component nylon 12 polymer were applied to the spinning.

The resultant undrawn composite filaments had a smooth surface, and drawn filaments had excellent carding and needling properties under the conditions described in Example 2. Also, the web prepared from the filaments had no pills thereon, and the sea component was easily dissolved off by trichloroethylene at a similar velocity to that of polystyrene.

EXAMPLE 26

The procedure of Example 21 was repeated, except that the island component was polyethylene terephthalate and the cross-sectional profile of the resultant composite filament was as shown in Fig. 1G.

The resultant undrawn composite filaments had a smooth surface, and drawn filaments had excellent carding and needling properties

under the conditions described in Example 2. Also, the web prepared from the filaments had no pills thereon, and the sea component was easily dissolved off by trichloroethylene at a similar velocity to that of polystyrene.

EXAMPLE 27

The procedure of Example 22 was repeated, except that the island component was a polyethylene terephthalate type copolyester containing 5% by mol of isophthalic acid as an additional copolycondensation unit.

The resultant undrawn composite filaments had a smooth surface, and drawn filaments had excellent carding and needling properties under the conditions described in Example 2. Also, the belt sheet prepared from the filaments had no pills thereon, and the sea component was easily dissolved off by trichloroethylene at a similar velocity to that of polystyrene.

EXAMPLE 28

The procedure of Example 24 was repeated, except that 40 parts of island component polymeric mixture and 60 parts of sea component styrene-copolymer were applied to spinning.

The resultant undrawn composite filaments had a smooth surface, and drawn filaments had excellent carding and needling properties under the conditions described in Example 2. Also, the belt sheet prepared from the filaments had no pill thereon, and the sea component was easily dissolved off by trichloroethylene at a similar velocity as that of polystyrene.

EXAMPLE 29

The procedure of Example 22 was repeated, except that the island component was a polyethylene terephthalate type copolyester containing 5% by mol of adipic acid as an additional copolycondensation unit, and number of the island component filaments was 24.

The resultant undrawn composite filaments had a smooth surface, and drawn filaments had excellent carding and needling properties under the conditions described in Example 2. Also, the web prepared from the filaments had no pills thereon, and the sea component was easily dissolved off by trichloroethylene at a similar velocity to that of polystyrene.

EXAMPLE 30

The procedure of Example 21 was repeated, except that the island component was polypropylene, and 25 parts sea component styrene-copolymer and 75 parts of island component polypropylene were applied to spinning.

The resultant undrawn composite filaments had a smooth surface, and drawn filaments had excellent carding and needling properties under the conditions described in Example 2. Also, the belt sheet prepared from the filaments had no pills thereon, and the sea component was easily dissolved off by trichloroethylene at a similar velocity to that of polystyrene.

EXAMPLE 31

The procedure of Example 22 was repeated, except that the island component was high density polyethylene.

The resultant undrawn composite filaments had a smooth surface, and drawn filaments had excellent carding and needling properties under the conditions described in Example 2. Also, the felt sheet prepared from the filaments had no pills thereon, and the sea component was easily dissolved off by trichloroethylene at a similar velocity to that of polystyrene.

EXAMPLE 32

The procedure of Example 21 was repeated, except that the sea component was a mixture of 20 parts polystyrene and 80 parts copolymer of 72% of methyl methacrylate, 24% of styrene and 4% of acrylonitrile.

The resultant undrawn composite filaments had a smooth surface, and drawn filaments had excellent carding and needling properties under the conditions described in Example 2. Also, the felt sheet prepared from the filaments had no pills thereon, and the sea component was easily dissolved off by trichloroethylene at a similar velocity to that of polystyrene.

EXAMPLE 33

The procedure of Example 1 was repeated, except that the spinning apparatus was that for the conventional "bi-metal" type composite filament, and the filament obtained had a bi-metal type cross-sectional profile and a fineness of 3 deniers, and 50 parts first component nylon 6 polymer and 50 parts second component styrene-copolymer were applied to spinning.

The resultant undrawn composite filaments had a smooth surface, and drawn filaments had excellent carding and needling properties under the conditions described in Example 2. Also, the felt sheet prepared from the filaments had no pills thereon, and the sea component was easily dissolved off by trichloroethylene at a similar velocity to that of polystyrene.

EXAMPLE 34

The procedure of Example 2 was repeated, except that the spinning apparatus was that for the conventional "core-in-a-sheath" type composite filament, and the filament obtained had a cross-sectional profile in which a core was concentrically embedded in a sheath and

a fineness of 3 deniers, and 50 parts core component nylon 66 polymer and 50 parts sheath component styrene-copolymer were applied to spinning.

The resultant undrawn composite filaments had a smooth surface, and drawn filaments had excellent carding and needling properties under the conditions described in Example 2. Also, the felt sheet prepared from the filaments had no pills thereon, and the sea component was easily dissolved off by trichloroethylene at a similar velocity to that of polystyrene.

EXAMPLE 35

The procedure of Example 21 was repeated, except that the spinning apparatus was that for the conventional "core-in-a-sheath" type composite filament, and the filament obtained had a cross-sectional profile in which a core was eccentrically embedded in a sheath and a fineness of 4 deniers, and 60 parts core composite nylon 6 polymer and 40 parts sheath component styrene-copolymer were applied to spinning.

The resultant undrawn composite filaments had a smooth surface, and drawn filaments had excellent carding and needling properties under the conditions described in Example 2. Also, the felt sheet prepared from the filaments had no pills thereon, and the sea component was easily dissolved off by trichloroethylene at a similar velocity to that of polystyrene.

WHAT WE CLAIM IS:—

1. A synthetic composite filament comprising a first filamentary component consisting of a copolymer of styrene with acrylonitrile and/or methyl methacrylate or a mixture of such copolymers, and a second filamentary component consisting of a synthetic crystalline fibre-forming polymer, the first and second components extending along the length of the composite filament, respectively and being bonded together continuously along the length of the composite filament.

2. A synthetic composite filament according to claim 1, wherein the first component consists of acrylonitrile-styrene copolymers, methyl methacrylate-styrene copolymers or acrylonitrile-methyl methacrylate-styrene copolymers.

3. A synthetic composite filament according to claim 2, wherein the acrylonitrile-styrene copolymer consists of 10 to 40% by weight of acrylonitrile units and 60 to 90% by weight of styrene units.

4. A synthetic composite filament according to claim 3, wherein the acrylonitrile-styrene copolymer consists of 10 to 24% by weight of acrylonitrile units and 76 to 90% by weight of styrene units.

5. A synthetic composite filament according to claim 2, wherein the methyl methacrylate-styrene copolymer consists of 10 to 90% by weight of methyl methacrylate units and 10 to 90% by weight of styrene units.

6. A synthetic composite filament according to claim 5, wherein the methyl methacrylate-styrene copolymer consists of 50 to 90% by weight of methyl methacrylate units and 10 to 50% by weight of styrene units.

7. A synthetic composite filament according to claim 2, wherein the acrylonitrile-methyl methacrylate-styrene copolymer consists of at most of 10% by weight of acrylonitrile units, at least 10% by weight of styrene units and more than 10% by less than 90% by weight of methyl methacrylate units.

8. A synthetic composite filament according to claim 1, wherein the second component consists of a polyamide, a polyester, or a polyolefin or of a mixture of two or of all three of these polymers or of a mixture of polymers in which at least one of these polymers is the main constituent.

9. A synthetic composite filament according to claim 8, wherein the polyamide is selected from the group consisting of nylon 6, nylon 66, nylon 12, nylon 610, nylon 9, nylon 11, block polyetheramides, copolymers and mixtures of these polyamides.

10. A synthetic composite filament according to claim 8, wherein said polyester is selected from the linear polyesters consisting of; at least one acid polycondensation unit selected from the group consisting of terephthalic acid, isophthalic acid, phthalic acid, p-hydroxy benzoic acid, adipic acid, sebacic acid, trimellitic acid, pyromellitic acid and sodium sulfoisophthalic acid; and at least one glycol polycondensation unit selected from the group consisting of ethylene glycol, propylene glycol, butylene glycol, cyclohexane dimethanol and diethylene glycol, and mixtures thereof.

11. A synthetic composite filament according to claim 10, wherein said polyester is polyethylene terephthalate.

12. A synthetic composite filament according to claim 8, wherein said polyolefin is selected from the group consisting of polyethylene, polypropylene, or a copolymer of or a mixture of polyethylene and polypropylene.

13. A synthetic composite filament as claimed in claim 1, which is of the "islands-in-a-sea" type, and in which the second component comprises a plurality of micro-fine filamentary components which extend independently parallel to the longitudinal axis of the composite filament and are embedded in the first component.

14. A synthetic composite filament according to claim 12, wherein the number of micro-fine components is at least ten.

15. A synthetic composite filament substantially as hereinbefore described with reference to the accompanying drawings.

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Printed for Her Majesty's Stationery Office, by the Courier Press, Leamington Spa, 1972.
Published by The Patent Office, 25 Southampton Buildings, London, WC2A 1AY, from which copies may be obtained.

1263221 COMPLETE SPECIFICATION

2 SHEETS *This drawing is a reproduction of the Original on a reduced scale*

Sheet 1

Fig. 1A

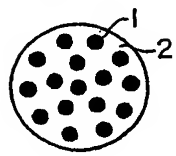

Fig. 1B

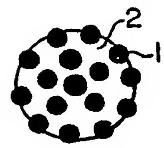

Fig. 1C

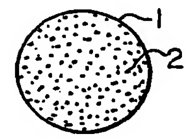

Fig. 1D

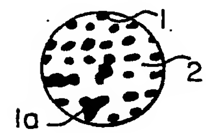

Fig. 2A

Fig. 2B

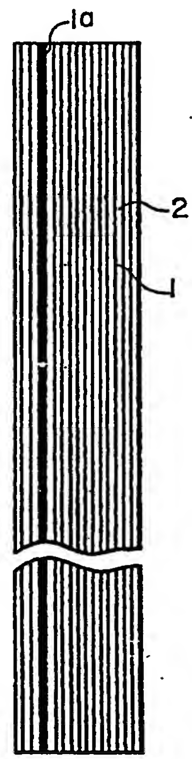

Fig. 1E

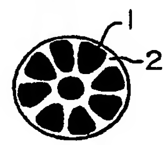

Fig. 1F

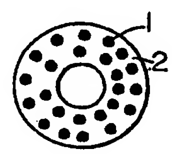

Fig. 1G

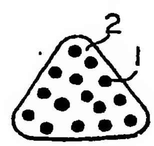

Fig. 1H

Fig. 3

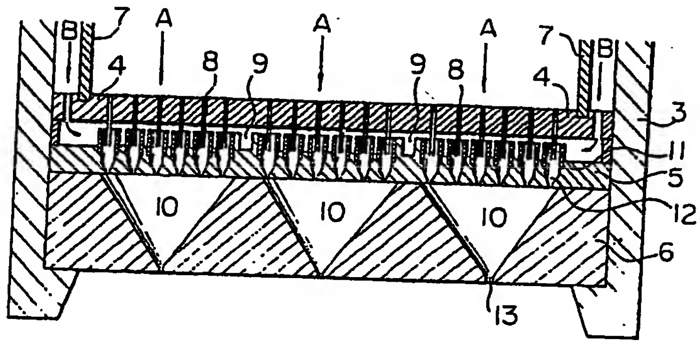
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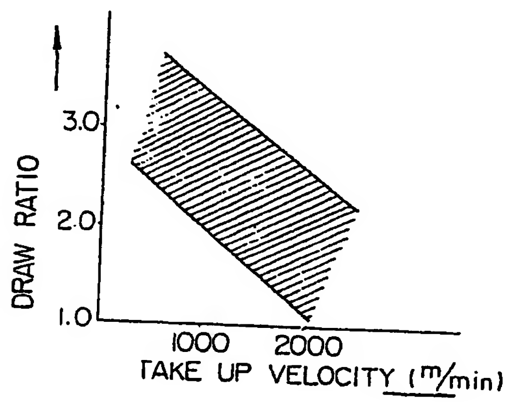
Fig. 4


DRAW RATIO
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